SCATTERING OF SLOW NEUTRONS BY CRYSTALLINE MATERIALS

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A short time after Max von Laue (1912) had discovered that X-rays are diffracted by crystals in similar ways as ordinary light is scattered by gratings, W. H. Bragg and W. L. Bragg developed a method to study the ordered arrangement of atoms in crystalline materials by using X-ray diffraction. Since then the technique has been vastly improved and extended to electron and neutron diffraction. In these investigations the main object was to determine the structure of the crystals and this is still an important subject in crystallography. Measurements are made of the intense elastic scattering, called Bragg scattering, which is due to interference between the waves scattered by the separate atoms. Careful analysis of measurements of scattering angles and relative intensities of the Bragg peaks give information concerning the arrangement of the atomic components in the crystal. Similar techniques have also been used to study the electron distribution in solids.

The dynamical properties of crystalline materials can be investigated by measuring the inelastic scattering. Use is then made of the fact that the incoming particle (photon, electron, neutron) can excite or deexcite eigen vibrations in the crystal (creation or annihilation of phonons) giving more or less direct information about these eigen states. The most clear-cut information is obtained from processes where only one phonon is created or annihilated. It is certainly possible to arrange the experiments in such a way that other effects are depressed, such as scattering with more than one phonon participating, multiple scattering, Compton scattering for X-rays etc., but in no case can they be completely neglected. When it became possible to produce intense beams of slow neutrons in neutron reactors, neutron diffraction became a very useful tool for studying e.g. lattice vibrations. The experimental technique is in principle the same for neutron and X-ray diffraction, but there are certain very important differences. Firstly, X-rays are mainly scattered by the electrons and the scattering intensity therefore rapidly decreases with decreasing atomic number, making it difficult to detect light atoms in X-ray experiments. In neutron diffraction the nuclei constitute the scatterers, at least in non-magnetic materials, and no such relation between intensity and atomic number as for X-rays exists. On the contrary the lightest atom, hydrogen, scatters neutrons exceptionally strongly. Owing to this fact neutrons are very useful for determining the structure of crystals containing light atoms. Neutron diffraction has also been used to study the magnetic arrangement

in, for instance, ferro-, antiferro- and ferri-magnetics. Here use is made of the fact that neutron scattering depends on the magnetic properties of the material owing to interaction between the magnetic moments of the neutron and the electrons in the atoms.

When considering inelastic scattering the main difference between X-rays and neutron diffraction is concerned with the energies. In both cases the wavelengths used are about 1Å. For the photons the corresponding energies are very much higher than the energies of the participating phonons (about 10^5 compared with unity) and the energy difference between the incoming and scattered photon is too small to be measurable. What is measured is therefore the scattering intensity in different directions. In the neutron case the energies of the neutrons are of the same order of magnitude as the energies of the phonons, and the energy exchange may be directly measured. When using cold neutrons these energies are even much lower than the energies of most of the participating phonons. Owing to this fact a more direct relation between quantities characterizing the eigen vibrations and the measurable quantities (energy exchange and intensity) is found than in the X-ray case. This is, however, counteracted by the fact that for neutrons the apparatus needed is much larger and the geometry poorer owing to lower intensity compared with available X-ray intensity. At present the accuracy of the results for the eigen vibrations obtained with X-rays and neutrons seems to be about the same.

The present dissertation is based on investigations published in the following papers:

I. On the scattering of slow neutrons by KCl, Arkiv för Fysik *13*, 199 (1958).
II. On two-phonon processes in neutron diffraction against crystals, Arkiv för Fysik *13*, 215 (1958).
III. Multi-phonon processes in slow neutron scattering by crystals, Arkiv för Fysik *14*, 315 (1958).

Several authors have previously stressed the possibility of using one-phonon scattering to investigate the eigen states of crystals and they have also given a general theory for these processes. It became clear that the inelastic scattering depends so sensitively on the details of the eigen vibrations that any comparison between theory and experiments must be based on more realistic assumptions about the lattice vibrations than for instance the Debye model. To investigate a realistic case the author therefore performed such a calculation for KCl (1955) using the numerical results from a theoretical investigation of the eigen vibrations by E. Tenerz. Our results, first communicated in mimeographed notes, are published in *paper I*. At the same time and independently similar calculations were performed by G. L. Squires for Al. His results show the same character as ours, giving a convincing proof that the details in the vibrational properties really manifest themselves in characteristic variations of the scattering intensity and energy, especially for cold neutrons. All the experiments which have now been made also confirm this statement.

In *paper II* two-phonon scattering is investigated to ascertain if the detailed properties of the eigen vibrations exert any significant influence also on two-phonon scattering. The same question had been raised earlier, but the investigations made were incomplete. We seem to be able to conclude, though no numerical calculations have been made, that in the coherent part the scattering really depends rather strongly on the details of the vibrations. The coherent scattering resembles the incoherent one-phonon scattering in some respects. We have also found a logarithmic singularity in the energy spectrum of the scattered neutrons for the same energies as obtained in one-phonon scattering. We wish to stress, however, that these peaks are in reality smoothed out owing to anharmonic effects in the crystal. The singularity may possibly manifest itself through a broadening of the one-phonon peaks and also a rather broad peak covering the one-phonon peaks especially for high temperatures.

A detailed study of the non-interferent part of the higher phonon scattering is made in *paper III*. The basic theory is developed by Akhieser, Pomeranchuk, Fröman, Van Hove and others. They do not give their results in a form convenient for direct numerical calculations, however, and it also seems hard to draw any detailed conclusions from the theory without further analysis of their expressions. The general character of the scattering is very different for low and high neutron energies as also for low and high temperatures. For low energies the main scattering originates from zero- and one-phonon scattering and the detailed properties of the crystal vibrations manifest themselves in characteristic features of the scattering. For large energies, on the other hand, the many-phonon scattering dominates, and it is then found that the neutrons are scattered by the crystal in approximately the same way as by a gas of free nuclei including the Doppler effect. After discussing this case intermediate energies are considered. The scattering cross section is expanded in the well-known phonon expansion, in which each term represents the scattering from processes where a fixed number of phonons participate. It is shown that the two- and higher phonon scattering mainly depends on two vibrational quantities, namely the mean kinetic energy per nucleus and the mean square deviation of the positions of the separate nuclei. The scattering thus only depends on some mean property of the crystal vibrations. We have further transformed the expression for the scattering cross section into a form convenient for numerical calculations by expanding the cross section in the Hermite orthogonal functions. Extensive numerical calculations have been carried out and these show that even in very accurate calculations only the first few terms in the expansion need be considered. The first term always gives the main part of the scattering and for many purposes this single term ought to give sufficient accuracy. In the last part of paper III the results obtained for the differential scattering cross section are used to derive approximate expressions for the total cross section, valid for cold neutrons but arbitrary temperatures and nuclei. There are several earlier investigations on the total cross section, most of them on the incoherent

part. In one of these G. Placzek suggests an expansion of the cross section in powers of m/M, m being the neutron mass and M the mass of a nucleus. He derives expressions which are very convenient for practical use. It is, however, difficult to see the range of validity of his formulae from his derivation. They cannot, for instance, be used for very light atoms and high temperatures. We have therefore considered Placzek's expansion from this point of view and have generalized his results in some respects.

The interference effects in three and higher phonon scattering have not been discussed. It can easily be shown that the terms involving many-phonons are very little influenced by these interference effects. It does not seem possible, however, to neglect them completely, at least not in three-phonon processes.

All the results in this dissertation are based on the harmonic approximation for the crystal vibrations, which implies that the phonons are considered to be independent of each other. This assumption ought to be realistic enough in many cases except for very high temperatures. It might, however, be a problem of current interest to investigate the corrections due to the anharmonic forces.

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